Simulating open quantum systems with Hamiltonian ensembles and the nonclassicality of the dynamics

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The main distinction between open and closed quantum systems is the intricate incoherent dynamics of the former, which is generically attributed to an ongoing correlation between the system and its environment. However, incoherent dynamics can also arise as a result of classical averaging over an ensemble of autonomous Hamiltonian evolutions, as it arises, e.g., in disordered quantum systems. Here, we discover a deeper correspondence between Hamiltonian ensembles and open quantum systems. We identify sufficient conditions under which a general system-environment Hamiltonian can be translated into a Hamiltonian ensemble on the system side. Moreover, we show how to construct an appropriate Hamiltonian ensemble to simulate the dynamics of the spin-boson model with arbitrary spectral density, even though the model Hamiltonian does not enjoy the ensemble form. The presence of an external driving, on the other hand, can destroy the validity of the Hamiltonian ensemble representation. This leads us to proposing a new way to witness the "nonclassicality" of open system evolutions.

Introduction.—When a quantum system is in contact with its environment, its dynamical behavior will in general deviate from the dynamics of a strictly isolated one [1–5]. As a result of an ongoing bipartite correlation arising from the interaction between the system and its environment, the system dynamics may display incoherent characteristics, such as dephasing or damping processes. Formally, such processes are captured by quantum master equations, replacing the von Neumann equation for isolated systems.

However, incoherent dynamics can also arise as a consequence of a purely classical averaging procedure over distinct autonomous evolutions. For example, the double slit experiment can, when exposed to a disordered potential and after averaging, encounter similar decoherence as if which-slit information had leaked into an environment [6]. In this sense, disordered quantum systems described by Hamiltonian ensembles can behave in an analogous manner as open quantum systems, even if individual realizations are strictly isolated.

Due to the dynamical analogy between open quantum systems and Hamiltonian ensembles, it has been shown [6, 7] to be fruitful to treat also the latter in terms of quantum master equations. However, their analogy is not restricted to this formal correspondence, but can be extended and put on a physical basis.

We approach this issue by considering a general class of system-environment arrangements, identifying the sufficient conditions, under which the total Hamiltonian gives rise to a Hamiltonian ensemble decomposition when discarding the environment. A simple but instructive implementation of this situation can be pictured in quantum information science, where a controlled-NOT (CNOT) gate is applied to an isolated qubit pair. Under suitable conditions [8], the reduced dynamics of the target qubit (T qubit) will then be described by the corresponding mixture of unitary evolutions.

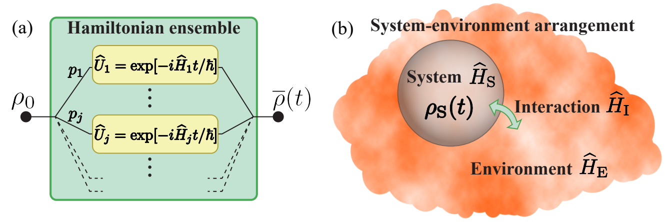

FIG. 1. (a) Schematic illustration showing the averaged state $\overline{\rho}(t)$ resulting from the Hamiltonian ensemble $\{(p_j, \widehat{H}_j)\}$. (b) We consider general system-environment arrangements and explore their correspondence with Hamiltonian ensembles by investigating the sufficient conditions leading to the ensemble-decomposition form.

To further explore the possibilities and limitations of this correspondence, we then proceed with the case where the conditions for the ensemble decomposition are not met. In particular, the Hamiltonian of the spin-boson model cannot be brought into the ensemble decomposition form. Interestingly, even if such decomposition is not possible, we prove that one is nevertheless able to construct an appropriate Hamiltonian ensemble, capable to simulate the full completely positive and trace-preserving (CPTP) dephasing dynamics. To one's surprise, this possibility to simulate is lost in the presence of an external driving.

By virtue of our findings, we strengthen the rela-

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tion between open systems and disordered quantum systems, and clarify under which circumstances a disordered quantum system can simulate an open system and vice versa. This may establish a new way to access the nonclassical nature of the dynamical processes, complementing, e.g., approaches based on the Wigner function [9] or the Glauber-Sudarshan P representation [10, 11]; moreover, it may pave the way towards identifying well-known consequences of disorder, such as, for instance, weak or strong localization, as originating from a system-environment coupling.

Dynamics of Hamiltonian ensembles.—An isolated quantum system is described by a Hamiltonian ensemble $\{(p_j, \widehat{H}_j)\}$, if the autonomous Hamiltonian $\widehat{H}_j$ of the system is drawn from a probability distribution p_j [see Fig. 1(a)]. This assumption, which underlies disordered quantum systems, may also be used, e.g., to describe experiments where the experimental apparatus is not fully controlled and the parameters are subject to variation. Usually one is then only interested in (or has access to) the ensemble state $\overline{\rho}(t)$, averaged over all possible evolutions.

The dynamics of the ensemble averaged state $\overline{\rho}(t)$ exhibits features distinct from any single realization, which is governed by the unitary evolution $\rho_j(t) = \widehat{U}_j\rho_0\widehat{U}_j^\dagger$, with the initial state ρ_0 and the unitary evolution operator $\widehat{U}_j = \exp[-i\widehat{H}_j t/\hbar]$; whereas the dynamics of the averaged state $\overline{\rho}(t)$ is given by the unital (i.e., identity invariant) map

$$\overline{\rho}(t) = \sum_j p_j e^{-\frac{i}{\hbar}\widehat{H}_j t}\rho_0 e^{\frac{i}{\hbar}\widehat{H}_j t}. \quad (1)$$

As one can show, an evolution equation for $\overline{\rho}(t)$ cannot be reduced to some effective Hamiltonian alone, but must in general take the form of a quantum master equation [6, 7].

A seminal and instructive example considers a single qubit subject to spectral disorder, i.e. the Hamiltonians in the ensemble only differ in their eigenvalues, while they share a common basis of eigenstates [7]. The Hamiltonian ensemble may then be given by $\{(p(\omega), \hbar\omega\hat{\sigma}_z/2)\}$, with the probability distribution $p(\omega)$ kept general. The resulting master equation in the Lindblad form reads

$$\frac{\partial}{\partial t}\overline{\rho}(t) = -\frac{i}{\hbar}[\varphi(t)\hat{\sigma}_z, \overline{\rho}(t)] + \gamma(t)[\hat{\sigma}_z\overline{\rho}(t)\hat{\sigma}_z - \overline{\rho}(t)], \quad (2)$$

where the effective energy $\varphi(t) = \hbar\mathrm{Im}[\partial_t \ln\phi(t)]/2$ and the decoherence rate $\gamma(t) = -\mathrm{Re}[\partial_t \ln\phi(t)]/2$ follow from the dephasing factor

$$\phi(t) = \int_{-\infty}^{\infty} p(\omega)e^{i\omega t}d\omega. \quad (3)$$

Depending on the underlying probability distribution $p(\omega)$, the master equation (2) can range from time-constant dephasing to a strongly oscillating incoherent behavior, the latter even giving rise to purity revivals [7].

It is worthwhile to recall that the occurrence of incoherent dynamics in the case of Hamiltonian ensembles is a consequence of the averaging procedure. Nevertheless, it is reminiscent of open quantum systems, where an ongoing correlation between system and environment gives rise to the incoherent dynamics. One objective of this article is to tighten this correspondence by exposing system-environment arrangements which result in Hamiltonian ensembles when tracing over the environment.

Environmentally-induced Hamiltonian ensembles.—A system-environment arrangement is characterized by a total Hamiltonian $\widehat{H}_{\mathrm{tot}} = \widehat{H}_{\mathrm{S}} + \widehat{H}_{\mathrm{E}} + \widehat{H}_{\mathrm{I}}$, with the system, $\widehat{H}_{\mathrm{S}}$, the environment, $\widehat{H}_{\mathrm{E}}$, and the interaction Hamiltonian $\widehat{H}_{\mathrm{I}}$ [see Fig. 1(b)]. The total system is typically assumed to evolve unitarily as $\rho_{\mathrm{tot}}(t) = \widehat{U}\rho_{\mathrm{tot},0}\widehat{U}^\dagger$, with $\widehat{U} = \exp[-i\widehat{H}_{\mathrm{tot}}t/\hbar]$. The reduced state of the system is then obtained by tracing over the environment: $\rho_{\mathrm{S}}(t) = \mathrm{Tr}_{\mathrm{E}}[\rho_{\mathrm{tot}}(t)]$.

We say that an open system is described by a Hamiltonian ensemble, if the system state $\rho_{\mathrm{S}}(t)$ allows a decomposition of the form (1), where the probabilities p_j and the Hamiltonians $\widehat{H}_j$ of the ensemble are determined by $\widehat{H}_{\mathrm{tot}}$ and the initial state $\rho_{\mathrm{tot},0}$ of the total system. Note that such a decomposition severely constrains the possible correlation between system and environment, as both p_j and $\widehat{H}_j$ are time-independent. The nonexistence of such decomposition for arbitrary open systems can also be deduced from the unitality of the map (1), which keeps the maximally mixed state invariant, while, for instance, in the case of spontaneous decay of an atom, any initial state is driven towards the ground state.

We begin our analysis by characterizing circumstances under which the reduced system states of system-environment arrangements, described by $\widehat{H}_{\mathrm{tot}}$, can enjoy the decomposition form (1). To address this, we focus on interaction Hamiltonians of the form

$$\widehat{H}_{\mathrm{I}} = \frac{1}{E_0}\widehat{H}_{\mathrm{S,I}} \otimes \widehat{H}_{\mathrm{E,I}}, \quad (4)$$

where E_0 is introduced for dimensional reasons and may stand for a characteristic energy scale in the system. This structure is found in many natural and generic system-environment arrangements. While Eq. (4) is not of the most general form, our reasoning can straightforwardly be carried over to interaction Hamiltonians with the structure $\widehat{H}_{\mathrm{I}} = \frac{1}{E_0}\sum_\mu \widehat{H}_{\mathrm{S,I},\mu} \otimes \widehat{H}_{\mathrm{E,I},\mu}$.

In practical scenarios, problems are often treated in the weak-coupling limit. This assumption ensures that perturbative treatments [12–15] are reliable and that the state of the environment is static at all times,

$$\rho_{\mathrm{E}}(t) = \mathrm{Tr}_{\mathrm{S}}[\rho_{\mathrm{tot}}(t)] = \rho_{\mathrm{E}}, \quad (5)$$

with $[\rho_{\mathrm{E}}, \widehat{H}_{\mathrm{E}}] = 0$. This static-environment assumption (SEA) is natural for many open systems and is thus widely adopted in open system theory. The SEA is weaker than the conventional Born approximation, which

explicitly eliminates all system-environment correlations, whereas the SEA still allows significant correlations. This will become clear below.

Hereafter, we further assume that $[\widehat{H}_{\mathrm{E}}, \widehat{H}_{\mathrm{E,I}}] = 0$. This assumption is distinct from the SEA (5), since the latter imposes constraints on the environmental state regardless of system-environment correlations, whereas the commutator requires a specific form of interactions. Therefore, the possible types of correlations are subject to limitations. Recently, a similar situation [16] and its experimental implementation in an all-optics setup [17] have been reported. Note that, if the SEA is replaced by the Born approximation, $\rho_{\mathrm{tot}}(t) = \rho_{\mathrm{S}}(t) \otimes \rho_{\mathrm{E}}$, the requirement $[\widehat{H}_{\mathrm{E}}, \widehat{H}_{\mathrm{E,I}}] = 0$ can be relaxed.

The time-evolved total state reads $\rho_{\mathrm{tot}}(t) = \widehat{U}_{\mathrm{S+I}}(\rho_{\mathrm{S},0} \otimes \rho_{\mathrm{E}})\widehat{U}^{\dagger}_{\mathrm{S+I}}$, with $\widehat{U}_{\mathrm{S+I}} = \exp[-i(\widehat{H}_{\mathrm{S}} + \widehat{H}_{\mathrm{I}})t/\hbar]$. Using the above commutators, we can reexpress the Hamiltonian in terms of an eigenbasis $\{|j\rangle_{\mathrm{E}}\}$ of $\widehat{H}_{\mathrm{E,I}}$ (i.e., $\widehat{H}_{\mathrm{E,I}} = \sum_j \varepsilon_{\mathrm{E,I},j} |j\rangle_{\mathrm{E}}\langle j|_{\mathrm{E}}$), which yields $\widehat{H}_{\mathrm{S}} + \widehat{H}_{\mathrm{I}} = \sum_j \widehat{H}_j \otimes |j\rangle_{\mathrm{E}}\langle j|_{\mathrm{E}}$, where

$$\widehat{H}_j = \widehat{H}_{\mathrm{S}} + \frac{\varepsilon_{\mathrm{E,I},j}}{E_0}\widehat{H}_{\mathrm{S,I}}. \tag{6}$$

We therefore have $\widehat{U}_{\mathrm{S+I}} = \sum_j \widehat{U}_j \otimes |j\rangle_{\mathrm{E}}\langle j|_{\mathrm{E}}$, with $\widehat{U}_j = \exp[-i\widehat{H}_j t/\hbar]$ and the time-evolved total state:

$$\rho_{\mathrm{tot}}(t) = \sum_j p_j e^{-\frac{i}{\hbar}\widehat{H}_j t} \rho_{\mathrm{S},0} e^{\frac{i}{\hbar}\widehat{H}_j t} \otimes |j\rangle_{\mathrm{E}}\langle j|_{\mathrm{E}}, \tag{7}$$

with probabilities $p_j = \langle j|_{\mathrm{E}}\rho_{\mathrm{E}}|j\rangle_{\mathrm{E}}$. If we now trace over the environment, $\rho_{\mathrm{S}}(t) = \sum_j \langle j|_{\mathrm{E}}\rho_{\mathrm{tot}}(t)|j\rangle_{\mathrm{E}}$, Eq. (7) reduces to the desired decomposition (1) in terms of the Hamiltonian ensemble $\{(p_j, \widehat{H}_j)\}$. We remark that in Eq. (7) the SEA is fulfilled, with the total state being exclusively classically correlated, displaying neither quantum discord [18, 19] nor entanglement. Consequently, the above two commutators, together with the initial direct product state, result, in case of interactions of the form (4), in classical system-environment correlations and a Hamiltonian ensemble decomposition.

As an instructive example, we consider a pair of qubits coupled to each other via a CNOT gate, where a control qubit (C qubit) determines the operation on a T qubit. If the state of the C qubit resides in a classical mixture $\rho^{\mathrm{C}} = a|1^{\mathrm{C}}\rangle\langle 1^{\mathrm{C}}| + (1-a)|0^{\mathrm{C}}\rangle\langle 0^{\mathrm{C}}|$ [8], the reduced dynamics of the T qubit will be described by the corresponding mixture of evolutions $\rho^{\mathrm{T}}(t) = a\widehat{U}_x\rho^{\mathrm{T}}_0\widehat{U}^{\dagger}_x + (1-a)\rho^{\mathrm{T}}_0$, with $\widehat{U}_x = \exp[-iJ\hat{\sigma}_x t/2\hbar]$ and J the coupling strength. In this case the C qubit plays the role of static environment and both above commutators vanish, as required.

Note that in general $[\widehat{H}_j, \widehat{H}_{j'}] \neq 0$. However, if $[\widehat{H}_{\mathrm{S}}, \widehat{H}_{\mathrm{S,I}}] = 0$, we obtain the case of global spectral disorder discussed in [7]; in the case of a single qubit as the system, this coincides with the general spectral disorder case (2). We emphasize that we did not have to make any assumptions on the system Hamiltonian $\widehat{H}_{\mathrm{S}}$.

Note that, in principle, any Hamiltonian ensemble $\{(p_j, \widehat{H}_j)\}$ can be generated from a suitably tailored system-environment arrangement. To see this, we write $\widehat{H}_j = \overline{H} + \widehat{V}_j$ (with the average $\widehat{\overline{H}} = \sum_j p_j\widehat{H}_j$) and assume that the interaction is of the form $\widehat{H}_{\mathrm{I}} = \sum_j \widehat{V}_j \otimes |j\rangle_{\mathrm{E}}\langle j|_{\mathrm{E}}$, i.e. it associates with each $\widehat{H}_j$ of the ensemble a distinct basis state $|j\rangle_{\mathrm{E}}$ of the environment. Taking the system Hamiltonian to be the average, $\widehat{H}_{\mathrm{S}} = \widehat{\overline{H}}$, and the environmental population as the probability distribution, $\rho_{\mathrm{E}} = \sum_j p_j|j\rangle_{\mathrm{E}}\langle j|_{\mathrm{E}}$, one recovers the desired arrangement under the SEA (5). Note that the index j is generic and may be continuous and/or a multi-index. The environment must then be chosen appropriately to accommodate the complexity of the Hamiltonian ensemble.

Simulating the spin-boson model.—After identifying conditions under which general arrangements may be interpreted in terms of Hamiltonian ensemble decompositions, we proceed with the spin-boson model described by

$$\hat{H}_{\mathrm{S}} = \frac{\hbar\omega_0}{2}\hat{\sigma}_z \qquad , \qquad \hat{H}_{\mathrm{E}} = \sum_{\vec{k}} \hbar\omega_{\vec{k}}\hat{b}^{\dagger}_{\vec{k}}\hat{b}_{\vec{k}},$$
$$\hat{H}_{\mathrm{I}} = \hat{\sigma}_z \otimes \sum_{\vec{k}} \hbar(g_{\vec{k}}\hat{b}^{\dagger}_{\vec{k}} + g^{*}_{\vec{k}}\hat{b}_{\vec{k}}). \tag{8}$$

The latter has been extensively studied and an analytical solution can be obtained [2]. Tracing over the environment, the system exhibits pure dephasing dynamics characterized by the dephasing factor

$$\phi(t) = \exp\left[i\omega_0 t - \Phi(t)\right]. \tag{9}$$

In contrast to Eq. (3), which results from averaging over a Hamiltonian ensemble, the dephasing factor (9) incorporates the information of the interaction and the environment into $\Phi(t) = 4\int_0^{\infty} \omega^{-2}\mathcal{J}(\omega)\coth\left(\hbar\omega/2k_{\mathrm{B}}T\right)(1-\cos\omega t)\,d\omega$, where $\mathcal{J}(\omega)$ is the environmental spectral density function. Whenever $\mathcal{J}(\omega)$ is given, the dynamical properties are specified. It should be emphasized that, for the solution above, the initial state is assumed to be in the direct product state. No further assumption is adopted for the analytical solution. Therefore, the total state is substantially entangled during the time evolution [20, 21].

We cannot employ the above reasoning to identify the ensemble decomposition form (6) for the spin-boson model (8), since $[\widehat{H}_{\mathrm{E}}, \widehat{H}_{\mathrm{E,I}}] \neq 0$. Instead, we now take a more general perspective and ask whether, given an arbitrary spectral density function $\mathcal{J}(\omega)$, one can nevertheless construct an appropriate Hamiltonian ensemble, with a valid probability distribution, that can (exactly) simulate the dynamics. In other words, we investigate the possibility of a Hamiltonian ensemble simulation, even if the actual reduced system state is not described by such.

To show that the answer is positive, we present a way to construct a Hamiltonian ensemble from the master equation. To this end, we must determine the member Hamiltonians and the corresponding probability distribution encapsulated in the ensemble. The master equation governing the pure dephasing dynamics is

$$\frac{\partial}{\partial t}\rho_{\mathrm{S}} = -i\left[\frac{\omega_0}{2}\hat{\sigma}_{\mathrm{z}}, \rho_{\mathrm{S}}\right] + \frac{1}{2}\frac{\partial \Phi(t)}{\partial t}\left(\hat{\sigma}_{\mathrm{z}}\rho_{\mathrm{S}}\hat{\sigma}_{\mathrm{z}} - \rho_{\mathrm{S}}\right). \quad (10)$$

In view of Eq. (2), we deduce that individual member Hamiltonians in the ensemble must be of the form $\omega\hat{\sigma}_{\mathrm{z}}/2$.

Given a probability distribution $p(\omega)$, the averaged dynamics can be determined by the Fourier transform according to Eq. (3). Conversely, the underlying distribution function leading to a specific dephasing factor (9) is obtained via the inverse transform

$$\wp(\omega) = \frac{1}{2\pi}\int_{-\infty}^{\infty} \exp[i\omega_0 t - \Phi(t)]\, \mathrm{e}^{-i\omega t} dt. \quad (11)$$

It is clear that the effect of ω_0 is merely to shift $\wp(\omega)$.

To be a legitimate probability distribution function, the resulting $\wp(\omega)$ in Eq. (11) must be normalized, $\int_{-\infty}^{\infty}\wp(\omega)d\omega = 1$, real, $\wp(\omega) \in \mathbb{R}, \forall\omega \in \mathbb{R}$, and positive, $\wp(\omega) \geq 0, \forall\omega \in \mathbb{R}$. Normalization is easily seen, since $\phi(0) = 1$ follows from the fact that the pure dephasing dynamics, characterized by the dephasing factor (9), should be CP. We therefore have $\int_{-\infty}^{\infty}\wp(\omega)d\omega = (2\pi)^{-1}\int_{-\infty}^{\infty}\exp\left[i\omega_0 t - \Phi(t)\right]2\pi\delta(t-0)dt = 1$. Moreover, since one is generically interested in the dynamical properties only for $t \geq 0$, we can deliberately extend the time domain to the full real axis such that $\Phi(t)$ is even and $\phi(-t) = \phi(t)^*$. This guarantees that $\wp(\omega)$ is real: $\wp(\omega) = (\pi)^{-1}\int_0^{\infty}\exp\left[-\Phi(t)\right]\cos(\omega-\omega_0)t dt \in \mathbb{R}$.

Positivity of $\wp(\omega)$ is less obvious, due to the sinusoidal factors of the integrand in Eq. (11). In the following, we invoke *Bochner's theory* [22] to prove the general positivity of $\wp(\omega)$. To this end, we first introduce the notion of positive definiteness. A function $f: \mathbb{R} \to \mathbb{C}$ is called to be positive definite if it satisfies $\sum_{j,k} f(t_j - t_k)z_j z_k^* \geq 0$ for any finite number of pairs $\{(t_j, z_j)|t_j \in \mathbb{R}, z_j \in \mathbb{C}\}$. Note that positive definiteness of a function is different from a positive function, since the latter may not necessarily be positive definite and vice versa. Rather, it corresponds to the positive semidefiniteness of a Hermitian matrix $[f(t_j - t_k)]_{j,k\in\mathcal{S}}$, formed by the function values $f(t_j - t_k)$ in accordance with a certain set of indices $\mathcal{S}$. As one can show, $\phi(t)$ in Eq. (9) is indeed positive definite. The proof is given in the Supplemental Material [23].

Bochner's theorem states, suitably expressed for our purposes, that a function f, defined on $\mathbb{R}$, is the Fourier transform of unique positive measure with density function $\wp$, if and only if f is continuous and positive definite [24, 25]. We can thus conclude that $\phi(t)$ in Eq. (9) is the Fourier transform of certain valid probability distribution $\wp(\omega)$, i.e. an analog to Eq. (3).

In summary, we have proven that, assuming that the pure dephasing dynamics (9) is CPTP, there exists a unique Hamiltonian ensemble $\{(\omega\hat{\sigma}_{\mathrm{z}}/2, \wp(\omega))\}$ which can simulate the dynamics of the spin-boson model exactly. In particular, Eq. (11) describes a legitimate, time-independent probability distribution for general $\phi(t)$, irrespective of the actual, possibly intricate, system-environment entanglement. This is one of our benchmark results.

External driving.—The possibility to simulate the spin-boson model with Hamiltonian ensembles appears natural from the view point of the unitality of the map (1). In fact, any pure dephasing dynamics is unital. One may thus conjecture that all CPTP pure dephasing dynamics correspond to certain Hamiltonian ensembles. Hereafter, we prove this conjecture wrong, by explicitly constructing a counterexample.

We again consider the spin-boson model with the system Hamiltonian replaced by a time-varying one, $\widehat{H}_{\mathrm{S}}(t) = \hbar\omega(t)\hat{\sigma}_{\mathrm{z}}/2$, while the environment Hamiltonian $\widehat{H}_{\mathrm{E}}$ and the interaction Hamiltonian $\widehat{H}_{\mathrm{I}}$ are kept intact as those in Eq. (8). The time dependence may be induced by applying an external driving field to the system. The driven spin-boson model also gives rise to pure dephasing dynamics of the reduced system. In this case, the dephasing factor is $\phi^{(\mathrm{D})}(t) = \exp\left[i\Omega(t) - \Phi(t)\right]$, where $\Omega(t) = \int_0^t \omega(\tau)d\tau$ and $\Phi(t)$ is same as the undriven one in Eq. (9).

Suppose that we apply an AC drive $\omega(t) = a\cos\omega_{\mathrm{d}}t$ with amplitude a and driving frequency ω_{d}. The resulting time-varying phase angle is $\Omega(t) = (a/\omega_{\mathrm{d}})\sin\omega_{\mathrm{d}}t$. With the help of Bessel's generating function, the distribution function in the presence of an AC drive becomes

$$\wp^{(\mathrm{D})}(\omega) = \sum_{n=-\infty}^{\infty} J_n\left(\frac{a}{\omega_{\mathrm{d}}}\right)\wp(\omega - n\omega_{\mathrm{d}}), \quad (12)$$

where $J_n(x)$ are Bessel functions and $\wp(\omega)$ is the distribution (11) in the absence of an external drive. As we show now, in the driven case $\wp^{(\mathrm{D})}(\omega)$ is, in general, not a positive distribution, even if the undriven $\wp(\omega)$ is a valid probability distribution.

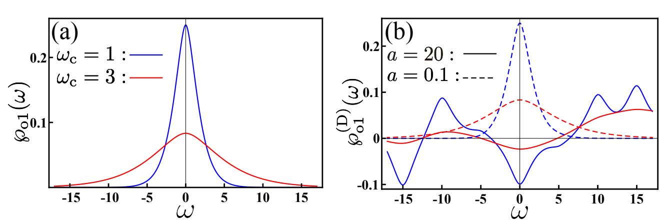

FIG. 2. (a) The distribution function $\wp_{\mathrm{o1}}(\omega)$, in the ensemble simulation of the undriven spin-boson model with the Ohmic spectral density, is a legitimate probability distribution function. (b) In the presence of an AC drive with amplitude $a = 20$ (solid curves), $\wp_{\mathrm{o1}}^{(\mathrm{D})}(\omega)$ may take negative values and thus no longer be a valid distribution function. However, when the AC drive is weak, with amplitude $a = 0.1$ (dashed curves), the validity of $\wp_{\mathrm{o1}}^{(\mathrm{D})}(\omega)$ still holds.

To demonstrate this, we consider the Ohmic spectral density $\mathcal{J}_{\rm o1}(\omega) = \omega \exp(-\omega/\omega_{\rm c})$. For simplicity, we further assume a degenerate system Hamiltonian (i.e., $\omega_0 = 0$) and the zero temperature limit. Then, the dephasing factor is $\phi_{\rm o1}(t) = \exp\left[-\Phi_{\rm o1}(t)\right]$, with $\Phi_{\rm o1}(t) = 2\ln\left(1 + \omega_{\rm c}^2 t^2\right)$ in the absence of an external drive.

In Fig. 2(a), we depict the distribution $\wp_{\rm o1}(\omega) = (4\omega_{\rm c}^2)^{-1}(\omega_{\rm c} + |\omega|)\exp\left(-|\omega|/\omega_{\rm c}\right)$ for $\omega_{\rm c} = 1$ (blue curve) and 3 (red curve) without drive. Note that $\wp_{\rm o1}(\omega)$ describes a legitimate probability distribution. Consequently, the Hamiltonian ensemble $\{(\omega\hat{\sigma}_{\rm z}/2, \wp_{\rm o1}(\omega))\}$ resembles the same pure dephasing dynamics characterized by $\phi_{\rm o1}(t)$. As expected, $\wp_{\rm o1}(\omega)$ is, due to the degeneracy of the system Hamiltonian, centered at $\omega = 0$, and broadens with increasing $\omega_{\rm c}$.

Applying AC driving, on the other hand, may result in an invalid distribution. In Fig. 2(b), a strong AC driving with $a = 20$ (solid curves) and $\omega_{\rm d} = 5$ leads to negative values of $\wp^{\rm (D)}(\omega)$ for both $\omega_{\rm c} = 1$ (blue curves) and 3 (red curves), violating positivity. However, when the AC driving is weak ($a = 0.1$, dashed curves), the validity of $\wp_{\rm o1}^{\rm (D)}(\omega)$ still holds.

Nonclassicality of the dynamics.—Hereafter, we suggest a physical interpretation of (quasi)distributions with negative values in terms of witnesses of nonclassicality of the system dynamics with reservoir interactions. This approach is distinct from other definitions of the nonclassicality of the dynamics focusing on the output states [26, 27]. In these approaches, the nonclassicality of the dynamics is traced back to the nonclassicality of the output states. Typically, the nonclassicality of the states is then captured by the Wigner function [9] or the Glauber-Sudarshan P representation [10, 11], which may become negative for a state configuration without classical counterpart [28–30]. Note, however, that, even though being genuine quantum states, coherent states are conceived to be classical-like, due to the positivity of their Wigner function.

For a total state of the form (7), the system-environment correlation is fully classical, neither displaying quantum discord nor entanglement at any later time. In such case, where the system dynamics (of an arrangement) can be simulated by a Hamiltonian ensemble, it is impossible to decide whether the dynamics follows from a system-environment interaction or such time-independent Hamiltonian ensemble. We therefore regard this case to be classical-like, as it refers to classical correlations. In contrast, if it is not possible to construct a suitable Hamiltonian ensemble to simulate the system dynamics, the latter must be governed by quantum correlations between the system and the environment. In such case, one may say the quantum correlations are witnessed by the impossibility to construct the simulation.

For the spin-boson model discussed above, the unital dynamical behavior is classical-like in the absence of an external drive, due to the existence of a Hamiltonian ensemble simulation. Applying a strong drive can break this possibility to simulate, witnessing the nonclassicality of the system-environment correlations.

Conclusions.—Inspired by the analogy between open quantum systems and Hamiltonian ensembles, we illuminated their correspondence from various perspectives. First, we considered a general class of system-environment arrangements and identified sufficient conditions, under which the total Hamiltonian gives rise to a Hamiltonian ensemble decomposition. While these conditions are not met in the spin-boson model, we nevertheless then constructed, on a more abstract level, an appropriate Hamiltonian ensemble to simulate its unital pure dephasing dynamics. We showed that this possibility to simulate is broken in the presence of a strong external drive. We propose that such existence/non-existence of a simulating Hamiltonian ensemble may establish a new way to assess the nonclassical nature of dynamical processes.

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SUPPLEMENTARY MATREIAL: SIMULATING OPEN QUANTUM SYSTEMS WITH HAMILTONIAN ENSEMBLES AND THE NONCLASSICALITY OF THE DYNAMICS

In this supplementary, we prove the positive definiteness of the dephasing factor $\phi(t) = \exp[i\omega_0 t - \Phi(t)]$. For textual completeness, we recall the definition of positive definiteness.

Definition 1 (Positive definiteness)**.** A function f defined on $\mathbb{R}$ is called to be positive definite if it satisfies

$$\sum_{j,k} f(t_j - t_k) z_j z_k^* \geq 0 \tag{13}$$

for any finite number of pairs $\{(t_j, z_j) | t_j \in \mathbb{R}, z_j \in \mathbb{C}\}$.

One of the critical results in our work, the positive definiteness of $\phi(t)$, is then stated as follows:

Theorem 2. Suppose that $\phi(t) = \exp[i\omega_0 t - \Phi(t)]$ defines a CPTP pure dephasing dynamics. If we further assume that $\Phi(t)$ is even and $\phi(-t) = \phi(t)^*$, then $\phi(t)$ defined on $\mathbb{R}$ is positive definite.

The fact that $\phi(t)$ defines a CPTP pure dephasing dynamics implies that $\phi(0) = 1$, $\Phi(0) = 0$, and $|\phi(t)| \leq \phi(0)$ for any $t > 0$. This means that the coherence of the system can never exceed its initial value. These properties will be frequently used in the following proof.

Proof. To simplify the problem, we first observe that the positive definiteness of $\phi(t)$ is equivalent to that of $\exp[-\Phi(t)]$, since

$$\begin{aligned}&\sum_{j,k} \phi(t_j - t_k) z_j z_k^* = \\ &\sum_{j,k} \exp[-\Phi(t_j - t_k)] \left(e^{i\omega_0 t_j} z_j\right) \left(e^{i\omega_0 t_k} z_k\right)^* .\end{aligned} \tag{14}$$

Correspondingly, we can assume that $\omega_0 = 0$ without loss of generality.

Since Eq. (13) must be valid for any number of pairs, we give the proof in an inductive manner.

In the case of only one pair (t_1, z_1), Eq. (13) is trivially satisfied. We therefore start with the case of two pairs. As stated in the main text, Eq. (13) is equivalent to the positive semidefiniteness of the Hermitian matrix:

$$\mathcal{M}^{(2)} = \begin{bmatrix} 1 & \exp[-\Phi(t_2 - t_1)] \\ \exp[-\Phi(t_1 - t_2)] & 1 \end{bmatrix}. \tag{15}$$

It is automatically satisfied according to the CPTP dynamics defined by $\phi(t)$.

We proceed to show the positive semidefiniteness of the Hermitian matrix

$$\mathcal{M}^{(3)} = \begin{bmatrix} 1 & e^{-\Phi_{2,1}} & e^{-\Phi_{3,1}} \\ e^{-\Phi_{1,2}} & 1 & e^{-\Phi_{3,2}} \\ e^{-\Phi_{1,3}} & e^{-\Phi_{2,3}} & 1 \end{bmatrix}, \tag{16}$$

for the case of three pairs. In the above matrix, and hereafter, the abbreviation $\Phi_{j,k} = \Phi(t_j - t_k)$ has been adopted. Since $\mathcal{M}^{(3)}$ is three dimensional, it is generically hard to write down an analytic expression for its three eigenvalues λ_μ. Nevertheless, analyzing its characteristic polynomial gives us substantial knowledge on the eigenvalues:

i) $\lambda_1 + \lambda_2 + \lambda_3 = 3 \geq 0$ follows from the invariance of the trace.

ii) $\lambda_1\lambda_2 + \lambda_2\lambda_3 + \lambda_3\lambda_1$ equals to the sum of all principal minors of $\mathcal{M}^{(3)}$ of order 2 and is consequently non-negative, since each principal minor is non-negative, following the positive semidefiniteness of $\mathcal{M}^{(2)}$.

iii) $\lambda_1\lambda_2\lambda_3 = \det\left(\mathcal{M}^{(3)}\right)$. The positivity of the product of eigenvalues is verified with the help of a simple geometric visualization shown in Fig. 3. Explicitly expanding the determinant leads to

$$\begin{aligned}\det\left(\mathcal{M}^{(3)}\right) &= \left(1 - \cos^2\theta\right) - \left(a^2 + b^2 - 2ab\cos\theta\right) \\ &= \sin^2\theta - c^2,\end{aligned} \tag{17}$$

with the notation $\cos\theta = \exp[-\Phi_{3,2}]$, $a = \exp[-\Phi_{2,1}]$, and $b = \exp[-\Phi_{3,1}]$. This can be interpreted in terms of a triangle with circumcircle (dashed circle) of diameter $2r$ less than 1. With the help of $c/\sin\theta = 2r$, the positivity of Eq. (17) and, consequently, of the product of eigenvalues is then inferred.

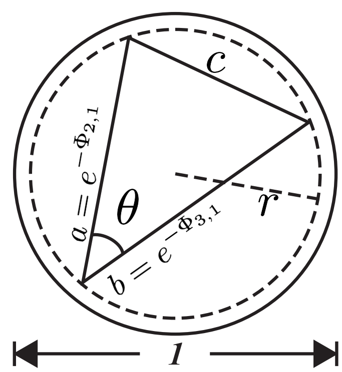

FIG. 3. A geometric visualization of Eq. (17). a and b can be considered as two sides of a triangle with angle θ and circumcircle (dashed circle) of diameter $2r$ less than 1. They are all enclosed in the circle (solid circle) of diameter 1.

Combining i)-iii), we can conclude that the three eigenvalues are non-negative each and, therefore, that $\mathcal{M}^{(3)}$ is positive semidefinite.

Before proceeding to the case of four pairs, it is worthwhile to discuss how the minimum of Eq. (13) is achieved. For the case of three pairs, the LHS of Eq. (13) is equiva-

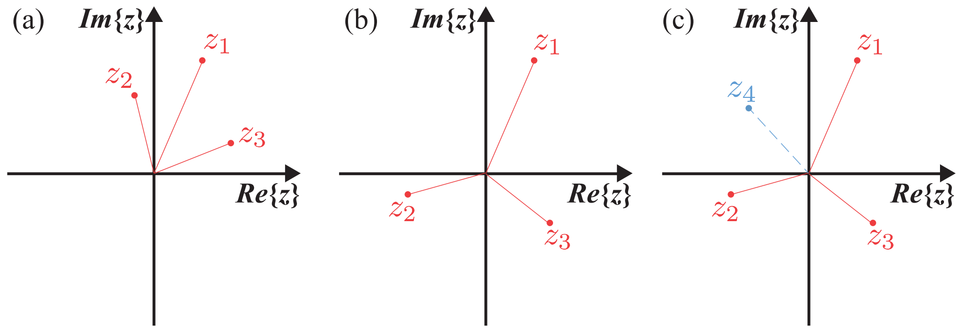

FIG. 4. (a) If the angles between any two z_j is less than $\pi/2$, the summation of all off-diagonal elements in array (18) is positive. (b) To maximize the negative contributions of off-diagonal elements, we must choose appropriate z_j, such that all the relative arguments strictly exceed $\pi/2$. (c) In the case of four pairs, it is impossible to insert the fourth z_4 such that all relative arguments are strictly larger than $\pi/2$.

lent to the summation over entries in the following array:

$$\begin{array}{ccc} |z_1|^2 & e^{-\Phi_{2,1}} z_2 z_1^* & e^{-\Phi_{3,1}} z_3 z_1^* \\ e^{-\Phi_{1,2}} z_1 z_2^* & |z_2|^2 & e^{-\Phi_{3,2}} z_3 z_2^* \\ e^{-\Phi_{1,3}} z_1 z_3^* & e^{-\Phi_{2,3}} z_2 z_3^* & |z_3|^2 \end{array} . \tag{18}$$

If we first determine the amplitudes $|z_j|$ and adjust their arguments and t_j, it is clear that the diagonal elements in the array (18) are all positive and, to reduce the resulting summation, the possible negative contributions are given by the off-diagonal elements. If we choose three pairs such that the angles between any two z_j within them is less than $\pi/2$, as show in Fig. 4(a), the summation of all off-diagonal elements is positive. Therefore, we must choose appropriate pairs such that all their relative arguments strictly exceed $\pi/2$, as show in Fig. 4(b). To maximize the negative contributions, we assume $t_1 = t_2 = t_3$ and $\exp[-\Phi_{j,k}] = 1$. We therefore draw the conclusion that $\sum_{j,k} f(t_j - t_k) z_j z_k^* \geq |z_1 + z_2 + z_3|^2$ for the case of maximized relative arguments between three z_j.

However, in the case of four pairs, it is impossible to insert the fourth z_4 such that all relative arguments are strictly larger than $\pi/2$, as shown in Fig. 4(c). According to above discussion, to deal with the array of four pairs,

$$\begin{array}{cccc} |z_1|^2 & e^{-\Phi_{2,1}} z_2 z_1^* & e^{-\Phi_{3,1}} z_3 z_1^* & e^{-\Phi_{4,1}} z_4 z_1^* \\ e^{-\Phi_{1,2}} z_1 z_2^* & |z_2|^2 & e^{-\Phi_{3,2}} z_3 z_2^* & e^{-\Phi_{4,2}} z_4 z_2^* \\ e^{-\Phi_{1,3}} z_1 z_3^* & e^{-\Phi_{2,3}} z_2 z_3^* & |z_3|^2 & e^{-\Phi_{4,3}} z_4 z_3^* \\ e^{-\Phi_{1,4}} z_1 z_4^* & e^{-\Phi_{2,4}} z_2 z_4^* & e^{-\Phi_{3,4}} z_3 z_4^* & |z_4|^2 \end{array} , \tag{19}$$

we can at most group three z_j with all three relative arguments strictly larger than $\pi/2$ by setting their corresponding t_j equal. Then the array (19) reduces to a simpler one:

$$\begin{array}{cc} |z_1 + z_2 + z_3|^2 & e^{-\Phi_{4,1}} z_4 (z_1 + z_2 + z_3)^* \\ e^{-\Phi_{1,4}} (z_1 + z_2 + z_3) z_4^* & |z_4|^2 \end{array} . \tag{20}$$

Again, in accordance with the positive semidefiniteness of $\mathcal{M}^{(2)}$, we can guarantee the validity of Eq. (13) for the case of four pairs.

For the case of five or more pairs, a similar procedure can be applied to continuously reduce the problem to an equivalent $\mathcal{M}^{(2)}$ or $\mathcal{M}^{(3)}$ case. This implies the validity of Eq. (13) for the general case. QED

We conclude by remarking that the proof of the positive definiteness of $\phi(t) = \exp[i\omega_0 t - \Phi(t)]$ already indicates the general impossibility of a Hamiltonian ensemble description in the driven case $\phi^{(\mathrm{D})}(t) = \exp[i\Omega(t) - \Phi(t)]$. Many conclusions in the above proof hold since the phase angle of $\phi(t)$ is directly proportional to time t. This is particularly manifest in Eq. (14). However, in general this is not the case, especially not in the strongly-driven case. We consequently may obtain invalid (or quasi-) distributions in the spin-boson model with strong driving.